

Dipole emission and coherent transport in random media I

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This is the first of a series of papers devoted to develop a microscopical approach to the dipole emission process and its relation to coherent transport in random media. In this Letter, we deduce general expressions for the decay rate of spontaneous emitters and the power emission of induced dipoles embedded in homogenous dielectric media. We derive formulae which apply generically to virtual cavities and, in the continuum approximation, to small real cavities.

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It is a general issue in physics the characterization of a system by means of its coherent transport properties and the study of the decay of unstable local states. In the quite general case that the constituents of the medium couple to each other through dipole-dipole interactions, the decay of an excited particle takes place through radiative and non-radiative emission. In particular, it is known that the spontaneous emission rate, Γ , in a dielectric medium depends on the interaction of the emitter with the environment [1]. This is so because the surrounding medium determines the number of channels into which the excited particle can decay. That is, the local density of states (LDOS). It is in this sense that the net effect of the random medium is to renormalize the vacuum as seen by the emitter. On the other hand, LDOS and Γ depend, upon additional properties of the emitter, on the statistical parameters which determine the coherent transport features of the medium. That is, on the electric susceptibility $\bar{\chi}$, the refraction index and the mean free path. As a matter of fact, null transmittance and inhibition of spontaneous emission are expected to occur in photonic band gap materials [2]. The understanding of life-times in random medium is relevant in the context of fluorescence biological imaging [3] and nano-antennas [4]. On the other hand, understanding of unconventional coherent transport properties is essential in engineering metamaterials for electromagnetic and acoustic waves [5]. In this Letter we deduce general formulae for the spontaneous and induced dipole power emission in a homogeneous random medium characterized by its electrical susceptibility. Longitudinal and transverse components are differentiated. It paves the way for the deduction of the relation between LDOS and Γ with coherent transport parameters.

The power W_ω^μ emitted in the process of spontaneous decay of a point dipole from an excited state $\Psi(\omega)$ is directly proportional to its decay rate Γ_ω^μ . The relation is given by $\Gamma_\omega^\mu = \frac{4}{\omega \hbar \epsilon_0} W_\omega^\mu$, where

$$\begin{aligned} W_\omega^\mu &= \frac{\omega \epsilon_0}{2} \Im \{ \vec{\mu} \cdot \vec{E}_{exc}^* \} = \frac{\omega^3}{2c^2} \Im \{ \vec{\mu} \cdot \bar{\mathcal{G}}_\omega^*(\vec{r}, \vec{r}) \cdot \vec{\mu}^* \} \\ &= -\frac{\omega^3}{6c^2} |\mu|^2 \Im \{ \text{Tr} \{ \bar{\mathcal{G}}_\omega(\vec{r}, \vec{r}) \} \}. \end{aligned} \quad (1)$$

In the above equation, Tr is the trace operator, μ is the dipole transition amplitude, ω is the transition frequency and $\bar{\mathcal{G}}_\omega(\vec{r}, \vec{r})$ is the propagator of the field emitted by the dipole in the process back to itself. We denote 2-rank tensors with overlines. In general, μ , ω and $\bar{\mathcal{G}}_\omega$ all depend on the interaction between the emitter and the host medium. On the other hand, LDOS is proportional to $\Im \{ \text{Tr} \{ \bar{\mathcal{G}}_\omega(\vec{r}, \vec{r}) \} \}$ and so are Γ_ω^μ and W_ω^μ [6].

Next, consider that the emitter consists of the dipole induced by a fixed exciting field $\vec{E}_{exc}(\vec{r}) = \vec{E}_0^\omega(\vec{r})$ on a scatterer of radius a and dielectric contrast $\epsilon_e(\omega)$. ω is assumed far from any resonance frequency of the emitter. Smallness implies $a \ll k_0^{-1}$, $k_0 = \omega/c$ being the bare wave number. Thus, $\vec{E}_0^\omega(\vec{r})$ is uniform within the scatterer. If the emitter is one of the host scatterers in a homogeneous and isotropic random medium, the emitted power reads

$$\begin{aligned} W_\omega^\alpha &= \frac{\omega \epsilon_0^2}{2} \Im \left\{ \int d^3r \chi_e^\omega \Theta(r-a) \int d^3r' d^3r'' \bar{\mathcal{G}}_\omega(\vec{r}, \vec{r}') \right. \\ &\quad \cdot [\bar{G}^{(0)}]^{-1}(\vec{r}', \vec{r}'') \cdot \vec{E}_0^\omega(\vec{r}'') \cdot \vec{E}_0^{*\omega}(\vec{r}) \left. \right\}, \end{aligned} \quad (2)$$

where $\chi_e^\omega = (\epsilon_e(\omega) - 1)$ –not to be confused with the susceptibility of the random medium– and

$$\bar{\mathcal{G}}_\omega(\vec{r}) = \bar{G}^{(0)}(\vec{r}) \sum_{m=0}^{\infty} \left[-k_0^2 \chi_e^\omega \int \Theta(v-a) \bar{\mathcal{G}}_\omega(v) d^3v \right]^m \quad (3)$$

$\bar{G}^{(0)}(\vec{r})$ being the tensor propagator of the electric field in free space. In spatial space representation, it consists of a Coulombian field propagator $\bar{G}_{Co.}^{(0)}(r) = \left[\frac{1}{k_0^2} \vec{\nabla} \otimes \vec{\nabla} \right] \left(\frac{-1}{4\pi r} \right)$ plus a radiation field propagator, $\bar{G}_{rad.}^{(0)}(r) = \frac{e^{i\omega r}}{4\pi r} \mathbb{I} + \left[\frac{1}{k_0^2} \vec{\nabla} \otimes \vec{\nabla} \right] \frac{e^{i\omega r} - 1}{-4\pi r}$. In Eq.(2), $\bar{\mathcal{G}}_\omega(\vec{r}, \vec{r}')$ is the propagator of the field emitted at a point \vec{r}' inside the particle back to another point \vec{r} also within the particle. Notice that, because the emitter is polarizable in this case, the wave comes back and forth infinite times with propagator $\bar{\mathcal{G}}_\omega$. Hence, series Eq.(3). In the above equation, for $a \ll k_0^{-1}$, the electric field is nearly uniform and we can approximate $\int \Theta(r-a) \bar{\mathcal{G}}_\omega(r) \simeq \frac{4\pi}{3} a^3 \bar{\mathcal{G}}_\omega(0)$. This expression is formally correct. However, the perturbative expansion of $\bar{\mathcal{G}}_\omega(0)$ contains singularities hidden in both

the electrostatic and radiative parts of $\bar{G}^{(0)}$. The later is related to resonance frequencies of the emitter polarizability [7] and so, negligible in the present case. The former is regularized by considering the finite size of the emitter, $\text{Lim}\{\int d^3r \Theta(r-a)\bar{G}^{(0)}(r)\} = \frac{1}{3k_0^2}\mathbb{I}$ as $a \rightarrow 0$. One way to avoid carrying this limit in further calculations consists of dressing up the single particle susceptibility with all the 'in-vacuum' electrostatic corrections at once. That is, by defining $\tilde{\chi}_e^\omega \equiv \frac{3}{\epsilon_e+2}\chi_e^\omega$ and the electrostatic polarizability $\alpha_0 \equiv 4\pi a^3 \frac{\epsilon_e-1}{\epsilon_e+2}$. On the other hand, the part free of singularities can be written as

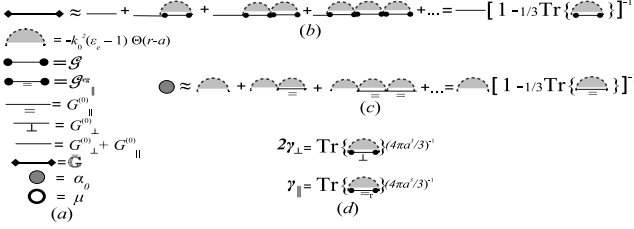


FIG. 1: (a) Feynman's rules. (b) Diagrammatic representation of Eq.(3). (c) Diagrammatic representation of the dressing up of χ_e^ω leading to α_0 . Approximation symbols denote that the field within the emitter is taken uniform. (d) Diagrammatic representation of Eq.(4).

$$\begin{aligned} \bar{\mathcal{G}}_\omega^{reg}(0) &= \frac{1}{3} \left[\int \frac{d^3k}{(2\pi)^3} 2\mathcal{G}_\perp^{reg}(k) + \int \frac{d^3k}{(2\pi)^3} \mathcal{G}_\parallel^{reg}(k) \right] \mathbb{I} \\ &\equiv \frac{1}{3} [2\gamma_\perp + \gamma_\parallel] \mathbb{I}, \end{aligned} \quad (4)$$

where $\mathcal{G}_{\perp,\parallel}^{reg}(k)$ are singularity-free and we have defined the $\gamma_{\perp,\parallel}$ factors in the last row. In Eq.(4), the scripts \parallel and \perp denote, in Fourier space, the tensor components along and transverse to the propagation direction respectively. Note that longitudinal and transverse modes couple to each other in the series Eq.(3) as scattering takes place off the dipole surface. With the above definitions we can rewrite Eq.(2) in terms of electrostatically renormalized operators – compare with [7, 8],

$$\begin{aligned} W_\omega^\alpha &= \frac{\omega\epsilon_0^2}{2} \Im \left\{ \int d^3r \tilde{\chi}_e^\omega \Theta(r-a) \int d^3r' d^3r'' \bar{\mathcal{G}}_\omega(\vec{r}, \vec{r}') \right. \\ &\quad \cdot [\bar{G}^{(0)}]^{-1}(\vec{r}', \vec{r}'') \cdot \vec{E}_0^\omega(\vec{r}'') \cdot \vec{E}_0^{\omega*}(\vec{r}) \left. \right\}, \end{aligned} \quad (5)$$

where

$$\bar{\mathcal{G}}_\omega(\vec{r}, \vec{r}') \equiv \bar{G}^{(0)}(\vec{r}, \vec{r}') \sum_{m=0}^{\infty} (-k_0^2 \alpha_0)^m 3^{-m} (2\gamma_\perp + \gamma_\parallel)^m.$$

The power emitted/absorbed by the induced dipole ac-

cording to Eq.(5) is

$$W_\omega^\alpha = \frac{\omega\epsilon_0^2}{2} \Im \left\{ \frac{\alpha_0}{1 + \frac{1}{3}k_0^2\alpha_0[2\gamma_\perp + \gamma_\parallel]} \right\} |E_0^\omega|^2 \quad (6)$$

$$\begin{aligned} &= \frac{-\omega^3\epsilon_0^2}{6c^2} \left\{ \frac{|\alpha_0|^2}{|1 + \frac{1}{3}k_0^2\alpha_0[2\gamma_\perp + \gamma_\parallel]|^2} \Im\{2\gamma_\perp + \gamma_\parallel\} \right. \\ &\quad \left. - \frac{\Im\{\alpha_0\}}{|1 + \frac{1}{3}k_0^2\alpha_0[2\gamma_\perp + \gamma_\parallel]|^2} \right\} |E_0^\omega|^2. \end{aligned} \quad (8)$$

The term in Eq.(8) corresponds to the power absorbed within the emitter. The term in Eq.(7) corresponds to the power radiated into the medium. The latter contains contributions of both coherent and incoherent radiation together with power absorbed by the host scatterers.

Finally, consider the spontaneous emission of a point dipole like that in Eq.(1), but now with an additional in-vacuum polarizability $\alpha = \alpha_0[1 - ik_0^3\alpha_0/(6\pi)]^{-1}$. The situation is analogous to that of a fluorescent particle placed on top of a host scatterer. Thus, the dipole moment of the system emitter-host-particle reads $\vec{p} = \vec{\mu} + \frac{\omega^2}{c^2} \tilde{\alpha} \bar{\mathcal{G}}_\omega(0) \vec{\mu}$, $\tilde{\alpha}$ being the renormalized polarizability of the host particle. The spontaneous field emitted in the decay process gives rise to an induced dipole moment in the host particle which modifies the decay rate of the emitter. It is plain from the perturbative development in Fig.2 that the emitted power can be written as

$$\begin{aligned} W_\omega^{\alpha,\mu} &= \frac{\omega\epsilon_0^2}{2} \frac{|\mu|^2}{\epsilon_0^2} \Im \left\{ \alpha_0^{-2} \left[\frac{\alpha_0}{1 + \frac{1}{3}k_0^2\alpha_0[2\gamma_\perp + \gamma_\parallel]} - \alpha_0 \right] \right\} \\ &= \frac{-\omega^3}{6c^2} \frac{|\mu|^2}{|1 + \frac{1}{3}k_0^2\alpha_0[2\gamma_\perp + \gamma_\parallel]|^2} \left[\Im\{2\gamma_\perp + \gamma_\parallel\} \right. \\ &\quad \left. - \frac{k_0^2}{3} \Im\{\alpha_0\} |2\gamma_\perp + \gamma_\parallel|^2 \right], \end{aligned} \quad (9)$$

where we recognize again the power absorbed within the induced dipole in the last term and the power radiated into the surrounding medium in the remaining.

Emission in vacuum is obtained by setting $\bar{\mathcal{G}}_\omega(r) =$

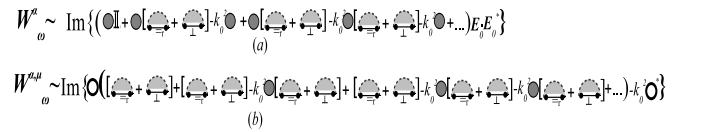


FIG. 2: (a) Diagrammatic representation of Eq.(2). (b) Diagrammatic representation of Eqs.(9,10).

$\bar{G}^{(0)}(r)$ in all the equations above.

For a complete description of W , Γ and LDOS we are just left with the computation of $\bar{\mathcal{G}}_\omega(\vec{r}, \vec{r})$ and its trace components, $2\gamma_\perp$ and γ_\parallel in each case. $\bar{\mathcal{G}}_\omega(\vec{r}, \vec{r})$ is the propagator of the field throughout the bulk from the emitter back to itself. It includes multiple-scattering processes which are in general spatially correlated. Correlations are both those among host scatterers themselves

and those of host scatterers with the emitter. If the emitter does not perturb the statistical isotropy of the host medium, it occupies the center of a spherical cavity of radius R . On top of that, the infinite series in Eq.(3) amount to multiple self-polarization processes. It is convenient to formulate additional Feynman's rules to describe the cavity and the self-polarization cycles. Those are given in Fig.3. A self-polarization cycle carries a factor $[2\gamma_{\perp} + \gamma_{\parallel}] \frac{1}{3} \mathbb{I} = \int d^3r \bar{\mathcal{G}}_{\omega}^{reg}(r) \delta^{(3)}(\vec{r})$ in the perturbative expansions for W in place of $(\frac{4\pi a^3}{3})^{-1} \int d^3r \bar{\mathcal{G}}_{\omega}^{reg}(r) \Theta(r - a)$ –see Fig.4(c,d). The cavity gives rise to a negative correlation function $h_C = -\Theta(r - R)$.

In addition, we have to describe the field propagation throughout the bulk. Special attention is to be paid to the interaction between longitudinal and transverse modes. For simplicity let us assume the host medium is infinite. Therefore, no coupling to surface modes needs to be considered. A bulk propagator \bar{G}^{ω} and dielectric and susceptibility tensors, $\bar{\epsilon}^{\omega}$ and $\bar{\chi}^{\omega}$ can be unambiguously defined. \bar{G}^{ω} is the dyadic Green's function of the macroscopic Maxwell equations for the time-mode ω in the bulk. We drop the script ω hereafter in all quantities. $\bar{\chi}$ carries correlation effects and is thus made of one-particle-irreducible (1PI) multiple-scattering events. In Fourier space, translation invariance and isotropy allow us to split Dyson equation for $\bar{G}(k)$ in two uncoupled and mutually orthogonal scalar algebraic equations,

$$G_{\perp}(k) = G_{\perp}^{(0)}(k) - k_0^2 G_{\perp}^{(0)}(k) \chi_{\perp}(k) G_{\perp}(k), \quad (11)$$

$$G_{\parallel}(k) = G_{\parallel}^{(0)}(k) - k_0^2 G_{\parallel}^{(0)}(k) \chi_{\parallel}(k) G_{\parallel}(k), \quad (12)$$

where $\bar{G}_{\perp}^{(0)}(k) = \frac{\Delta(\hat{k})}{k_0^2 - k^2}$ and $\bar{G}_{\parallel}^{(0)}(k) = \frac{\hat{k} \otimes \hat{k}}{k_0^2}$, \hat{k} being a unitary vector along the propagation direction and $\Delta(\hat{k}) \equiv I - \hat{k} \otimes \hat{k}$ being the projective tensor orthogonal to \hat{k} . The longitudinal component is the propagator of the electrostatic field. Likewise, the transverse component is the propagator of the radiation field. In view of Eqs.(11,12), longitudinal and transverse coherent photons do not couple to each other as traveling throughout a random medium as it is the case of photons in free space. Eqs.(11,12) can be solved independently yielding the renormalized propagator functions for the coherent –macroscopic– electric field, $\bar{G}_{\perp}(k) = \frac{\Delta(\hat{k})}{k_0^2 \epsilon_{\perp}(k) - k^2}$, $\bar{G}_{\parallel}(k) = \frac{\hat{k} \otimes \hat{k}}{k_0^2 \epsilon_{\parallel}(k)}$. However, a more detailed examen shows that longitudinal and transverse bare photons –i.e., normal modes of $\bar{G}^{(0)}$ – do couple necessarily when they propagate in a random medium and experience multiple scattering processes. In Eqs.(11,12), longitudinal and transverse bare photons enter both $\chi_{\perp}(k)$ and $\chi_{\parallel}(k)$ by means of the spatial correlations among scatterers. In particular, coupling between longitudinal and transverse modes shows up at the emitter cavity. We define the cavity

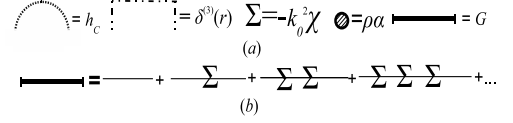


FIG. 3: (a) Feynman's rules. (b) Diagrammatic representation of the bulk propagator \bar{G} .

factors

$$\begin{aligned} C_{\perp}(k) &= \frac{1}{2} \int d^3r e^{i\vec{k} \cdot \vec{r}} h_C(r) \text{Tr}\{\bar{G}^{(0)}(r)[\bar{I} - \hat{k} \otimes \hat{k}]\} \\ &= \frac{1}{2} \int \frac{d^3k'}{(2\pi)^3} h_C(|\vec{k}' - \vec{k}|) \left[G_{\perp}^{(0)}(k') \right. \\ &\quad \left. + G_{\perp}^{(0)}(k') \cos^2 \theta + G_{\parallel}^{(0)}(k') \sin^2 \theta \right], \end{aligned} \quad (13)$$

$$\begin{aligned} C_{\parallel}(k) &= \int d^3r e^{i\vec{k} \cdot \vec{r}} h_C(r) \text{Tr}\{\bar{G}^{(0)}(r)[\hat{k} \otimes \hat{k}]\} \\ &= \int \frac{d^3k'}{(2\pi)^3} h_C(|\vec{k}' - \vec{k}|) \\ &\quad \times \left[G_{\parallel}^{(0)}(k') \cos^2 \theta + G_{\perp}^{(0)}(k') \sin^2 \theta \right], \end{aligned} \quad (14)$$

where $\cos \theta \equiv \hat{k} \cdot \hat{k}'$.

Consider first the emitter as either a spontaneous or induced dipole equivalent in all to the rest of host scatterers or as a fluorescent particle on top of a host scatterer. Therefore, it correlates to the surrounding as any host particle would do. The exclusion volume around the emitter is thus a virtual cavity as it does not perturb the medium at all. In each self-polarization cycle, the emitted/polarizing field experiences in the bulk multiple-scattering processes which are correlated to the emitter/receiver. A typical process of these is depicted in Fig.4(b). There, only 2-point correlation functions, $h(r)$, are used for simplicity. Note that, because the emitter and the receiver coincide, the correlations of any intermediate scattering process to one or the other extreme of the diagram are equivalent. This allows to attribute all the correlations to the emitter on the left, which amounts to the factor $\chi/\rho\alpha$ –Figs.4(c,d). Therefore, we have

$$2\gamma_{\perp} = \int \frac{d^3k}{(2\pi)^3} \left[\frac{2\chi_{\perp}(k)/(\rho\alpha)}{k_0^2[1 + \chi_{\perp}(k)] - k^2} \right] - 2\Re\{\gamma_{\perp}^{(0)}\} \quad (15)$$

$$\gamma_{\parallel} = \int \frac{d^3k}{(2\pi)^3} \left[\frac{1}{\rho\alpha} \frac{\chi_{\parallel}(k)}{k_0^2[1 + \chi_{\parallel}(k)]} - \frac{1}{k_0^2} \right], \quad (16)$$

where ρ is the density of host scatterers. The factor 2 in front of γ_{\perp} stands for the two transverse polarizations while there is only one longitudinal. The term $-2\Re\{\gamma_{\perp}^{(0)}\}$ accounts for the singularity of $\bar{G}_{rad}^{(0)}(0)$. This concludes the computation of the spontaneous emission of an emitter on top of a host scatterer as a function of the susceptibility tensor $\bar{\chi}$ of the host medium and the scatterer polarizability α_0 . We can split up the power emitted and the associated decay rates into longitudinal and

transverse components, $2\Gamma_{\perp}$ and Γ_{\parallel} , as it derives from Eq.(9) (compare with [8]). It is plain by substitution of $\bar{\chi}(k) \approx \rho\alpha - (\rho\alpha)^2 k_0^2 \bar{C}$ in Eqs.(11,12) that, for any $h_C(r) \neq \text{const}$, longitudinal and transverse photons do couple at the cavity surface. This is at the root of the contribution of longitudinal modes to the total decay rate, even in absence of absorbtion (see [12]). In particular, for $R \ll k_0^{-1}$, the only contribution to Eqs.(13,14) comes from longitudinal photons –i.e. electrostatics, yielding $C_{\parallel,\perp}(k) = \frac{-1}{3k_0^2}$. This is recognized as the Lorentz-Lorenz (LL) cavity factor [10].

Next, let us consider that the emitter is either an impurity itself or seats on top of a polarizable impurity within an arbitrary cavity. That breaks manifestly translation invariance. Thus, the cavity is real and it is not strictly possible to define a Dyson propagator. The reason being that, in any multiple scattering process, beside the translation-invariant correlation functions joining host scatterers, each scatterer i is correlated to the emitter through the 2-point correlation function $g_C(r_i) \equiv 1 + h_C(r_i)$. A first approximation to go around this problem can be made if $R \gg \xi$, ξ being the typical correlation length between host scatterers. In such a case we can consider that, for any 1PI diagram in $\bar{\chi}(\vec{r}_1, \vec{r}_2)$, the emitter gets correlated to any of the host scatterers therein by simple convolution of $\bar{\chi}(\vec{r}_1, \vec{r}_2)$ with $g_C(r_1)$. This yields the series expansion depicted in Fig.5(b) in which wave propagation still depends on the emitter location. A further approximation consists of considering that, for $k_0 R \ll 1$ the empty cavity do not affect the wave propagation but in its entrance and departure from the host medium. That is, both wave and emitter see a continuum beyond $r \sim R$. This approximation is closer to the Onsager–Böttcher (OB) [13] approach as reformulated in [14] (see also the quantum approach in [9]). If G is the bulk propagator as defined in absence of cavity, the γ functions take the form –see Fig.5(c),

$$2\gamma_{\perp} \simeq -i\frac{k_0}{2\pi} - 2k_0^2 \int \frac{d^3k}{(2\pi)^3} [C_{\perp} + G_{\perp}^{(0)}] \chi_{\perp} G_{\perp}^{(0)}(k) + 2k_0^4 \int \frac{d^3k}{(2\pi)^3} [C_{\perp} + G_{\perp}^{(0)}]^2 G_{\perp} \chi_{\perp}^2(k), \quad (17)$$

$$\gamma_{\parallel} \simeq -k_0^2 \int \frac{d^3k}{(2\pi)^3} [C_{\parallel} + G_{\parallel}^{(0)}] \chi_{\parallel} G_{\parallel}^{(0)}(k) + k_0^4 \int \frac{d^3k}{(2\pi)^3} [C_{\parallel} + G_{\parallel}^{(0)}]^2 G_{\parallel} \chi_{\parallel}^2(k), \quad (18)$$

where an analogous equivalence to that illustrated in Fig.4(b) has been used.

In summary, we have derived general expressions for the longitudinal and transverse components of the power emission and the decay rate of induced dipoles and spontaneous emitters respectively in function of the electrical susceptibility of the host medium. These are Eqs.(1;7,8;9,10), depending on the emitter nature,

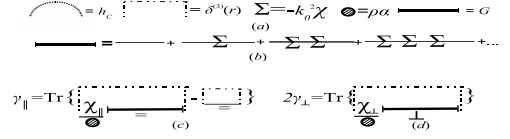


FIG. 4: (a) 2-point correlation function $h(r)$ and associated interaction vertex. (b) Diagrammatic representation of the equivalence between multiple-scattering processes amounting to \bar{G} . (c),(d) Alternative representation to Fig.1(d).

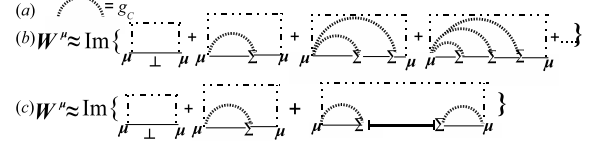


FIG. 5: (a) 2-point correlation function $g_c(r)$. (b) First approximation of W^μ . (c) Diagrammatic representation of the approximate formulas in Eqs.(17,18).

together with the appropriate γ -factors. Those are, Eqs.(15,16) for a source seated in a virtual cavity. They are exact and their diagrammatic representation is that of Fig.2(b) –up to constant prefactors. For a source seated in a real cavity drilled in a continuous medium, the γ -factors are in Eqs.(17,18). They are approximate and their diagrammatic representation is that of Fig.5(c).

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